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QUARTERLY REPORT

HYDROGEN-OXYGEN ELECTROLYTIC REGENERATIVE FUEL CELLS

by

M. Klein

prepared for

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

15 October 1966

CONTRACT NAS3-2781

Technical Management
NASA Lewis Research Center
Cleveland, Ohio
Auxiliary Power Generation Office
D. G. Soltis

ELECTRO-OPTICAL SYSTEMS, INC.
300 No. Halstead Street
Pasadena, California

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ILLUSTRATIONS

1. INTRODUCTION

This report reviews the progress made on the development of a regenerative hydrogen-oxygen fuel cell under Contract NAS3-2781 during the period of 1 July 1966 through 30 September 1966. The program objective is the development of an electrolytically regenerative, hydrogen-oxygen fuel cell that will be superior in performance to currently available rechargeable batteries. The device under development consists of a cell stack that is used as an electrolyzer during charge periods and as a fuel cell during discharge periods. Integral gas storage tanks are used to contain the hydrogen and oxygen generated during charge. Such a device offers advantages in the area of watt-hours-per-pound, high ambient temperature operation, and greater cycle life than that which can be obtained from existing secondary batteries. Phase I consisted of design, development, and testing of a nominal 75W, 44-watt-hour 6-cell unit to demonstrate the feasibility of a multicell regenerative device. This phase has been completed. Phase II (as modified) consists of investigation of the matrix, electrode combinations to improve cycle life, and the design and development of a 500W, 600-watt-hour, 34-cell unit of minimum weight for evaluation as a flight prototype.

2. SUMMARY

During this period primary emphasis was placed on the testing of single cells with various electrode and matrix structures to improve cycle life capabilities and to obtain a better understanding of modes of deterioration of cell performance. Single-cell life of 952 cycles was achieved with a 90 percent potassium titanate-10 percent asbestos matrix. This was the longest operational life achieved to date. Gradual degradation in performance is still observed; this may be caused by the 10 percent asbestos constituent. Attempts to substitute another inert fiber material for the asbestos have not proved successful as yet due to the development of cross gas leakage with these type mats. An alternate approach for the matrix which utilizes a gas barrier membrane sandwiched between two layers of absorbent material offers a possible solution to the recombination problem and thus may enable longer cycle life.

3. TECHNICAL DISCUSSION

3.1 Single Cell Tests

Twenty-nine single-cell tests were conducted during this period to evaluate the performance of various electrode and matrix structures. Test results and construction variables of these cells are summarized in Table I.

3.1.1 Ninety Percent Potassium Titanate (KT) – 10 Percent Asbestos (asb) Matrix Cell Test

During the preceding report period, testing was started on cell 198. The oxygen electrode of the cell was a gold-plated nickel screen platinized by electrodeposition of platinum (25 mg/cm^2) on the surface of the screen. The hydrogen electrode was a chemically platinized porous nickel plaque. The matrix was 90 percent KT and 10 percent asb.

As shown in Fig. 1, the cell was cycled continuously for a period of 952 cycles. During the cycling, there was a gradual increase in the charge voltage and a decrease in discharge voltage. After 952 cycles a short circuit developed within the cell, causing the cell voltage to drop to 0 during charge and discharge. The test was discontinued at this failure.

Since the load bank used to test the cells was a fixed resistor, the discharge current decreased as the cell voltage degraded during discharge. In the early cycling phases discharge current ranged from 15 to 18A, but as the voltage began to gradually drop during the latter cycling sequences, the current degraded steadily. At the start of the 952nd discharge cycle the current measured 15A; it dropped during discharge to 10A at the end of the cycle.

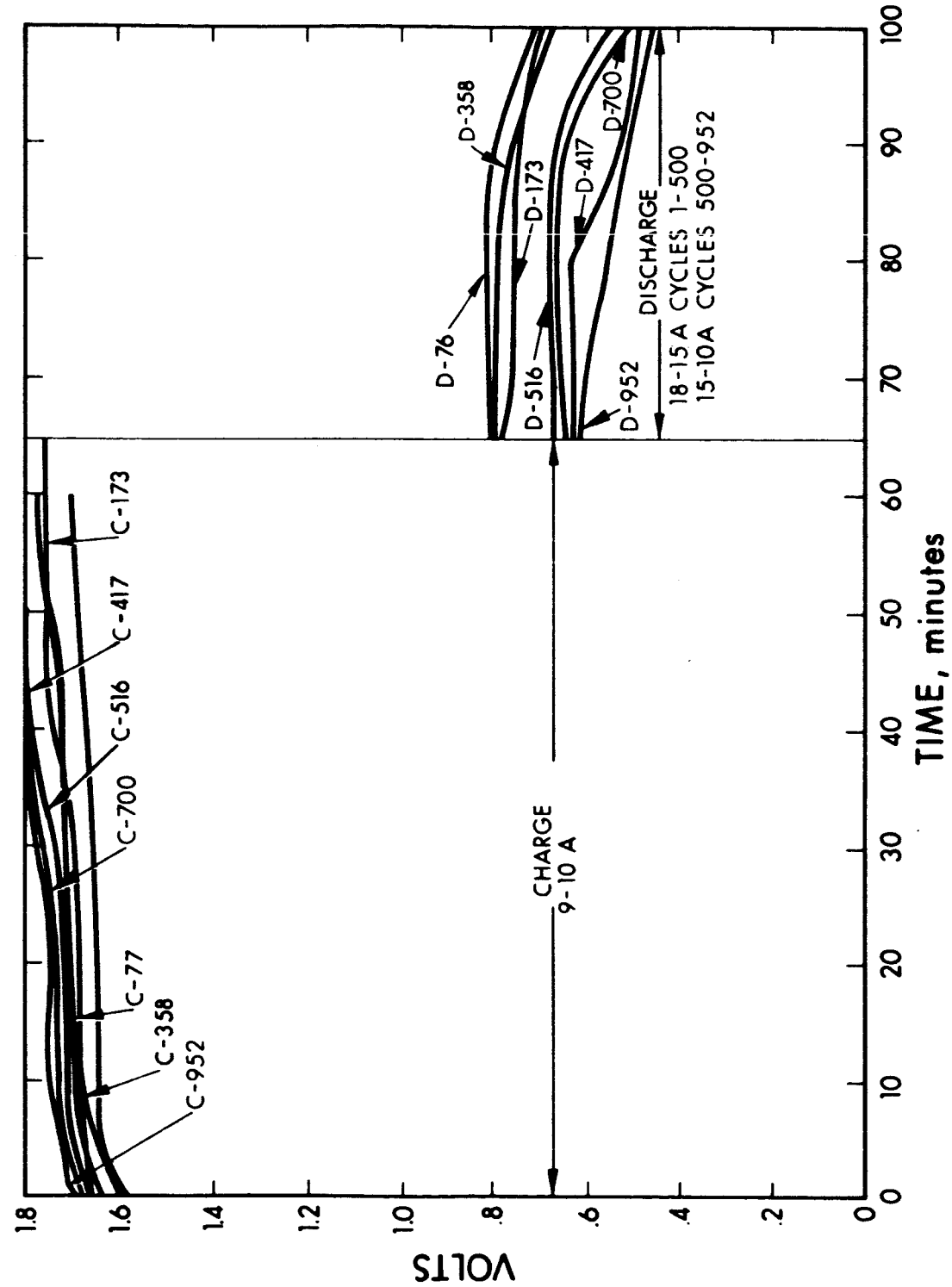

FIG. 1 CYCLING PERFORMANCE OF CELL 198

TABLE I

SUMMARY OF SINGLE-CELL TESTS

Cell No.	O_2 Electrode		H_2 Electrode		Matr Thick In. (%)	Matr Dry Wt (gm)	Comments*	Results
	Type	Catalyst	Type	Catalyst				
198	Plat Gold Screen	25 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	19.6		952 cycles, gradual degradation
199	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	24.5	O_2 electrodes from cell 196	Test stopped at 405 cycles, due to poor performance
202	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	20.09	O_2 electrodes from cell 195	387 cycles, cell developed internal short
205	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	25.0		52 cycles, developed slow recombination
206	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	20	O_2 electrode used in cells 196, 199	700 cycles gradual degradation
207	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	20	O_2 electrode from cell 202	25 cycles, developed slow recombination
208	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	24.5	Electrode from 6-cell 110	21 cycles. Good performance
209	Plat Ni Plaque	20 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	22.0	H_2 concentration cell	Discontinued test after 1415 hours final voltage 0.223
210	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	21.5	Electrodes from 6-cell 110	Ran poorly
211	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 ppl**	21.5		50 good cycles, then developed slow gas recombination

*Electrolyte, all cases: 40% KOH; 34 grams by weight unless otherwise noted

**ppl = polypropylene

TABLE I

SUMMARY OF SINGLE-CELL TESTS (contd)

Cell No.	O_2 Electrode		H_2 Electrode		Matr Thick In. (%)	Matr Dry Wt (gm)	Comments*	Results
	Type	Catalyst	Type	Catalyst				
212	Am Cy	40 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	22.0		418 cycles, then developed slow recombination last 17 cycles
213	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 ppl**	22.0	O_2 electrode was used in cell 211	119 cycles, then developed slow gas recombination
214	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	21.5	O_2 electrode used in cells 196,199,and 206	464 cycles, then developed slow gas recombination
215	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 ppl	21.5	Used electrodes	30 cycles, then developed slow gas recombination
216	Am Cy	9 mg Pt/cm^2	Am Cy	9 mg Pt/cm^2	90 KT 10 ppl	21.5	New electrodes	209 cycles; developed recombination
217	Am Cy	40 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	24.1	Used electrodes from cell 212	432 cycles; recombination
218	Plat Ni Plaque	20 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	21	Electrolyte wt: 35 gm. H_2 concentration cell	600 hours; still running
219	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	21.3	Electrolyte wt: 50 gm. No backup screens	Poor performance
220	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	0.060 asb	27.3	Electrolyte wt: 50 gm	High impedance cell

*Electrolyte, all cases: 40% KOH; 34 grams by weight unless otherwise noted

**ppl = polypropylene

TABLE I

SUMMARY OF SINGLE-CELL TESTS (contd)

Cell No.	O_2 Electrode		H_2 Electrode		Matr Thick In. (%)	Matr Dry Wt (gm)	Comments*	Results
	Type	Catalyst	Type	Catalyst				
221	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	0.060 asb	26.1	Electrolyte wt: 50 gm. No backup screens	Poor performance
222	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)13.6 (2)13	Two matrixes; Electrolyte: 50 gm. No backup screens. Matrix: membrane (polyethylene)	Ran poorly; 5 cycles
223	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 ppl**			40 cycles; recombination developed on cycle 22
224	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 ppl	21.4	Electrolyte: 30% KOH; 37 gm	Discontinued after 24 cycles; recombination developed
225	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	21.7	Electrolyte: 49% KOH; 37 gm. Used electrodes from 6-cell unit	10 cycles good. Matrix was subsequently damaged by H_2 leak
226	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)13.8 (2)14	Electrolyte: 40.3% KOH; 40 gm. Polyethylene membrane. New electrodes. 0.040 separator	4 cycles; poor performance

*
**Electrolyte, all cases: 40% KOH; 34 grams by weight unless otherwise noted
ppl = polypropylene

TABLE I

SUMMARY OF SINGLE-CELL TESTS (contd)

Cell No.	O_2 Electrode		H_2 Electrode		Matr Thick In. (%)	Matr Dry Wt (gm)	Comments*	Results
	Type	Catalyst	Type	Catalyst				
227	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)14.1 (2)13.6	Electrolyte: 40.3% KOH; 40 gm total (20 gm/ matrix). 0.5-inch spacer	Damaged matrix with large differential
228	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)13.2 (2)13.2	Matrix of woven Teflon cloth. Electrolyte: 40.3% KOH; 40 gm total (20 gm/ matrix). 0.5-inch spacer	39 cycles; poor performance
229	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)13.3 (2)13.5	Matrix: cellulose membrane (FSC) Electrolyte: 40.3% KOH; 40 gm (20 gm/matrix). 0.5-inch spacer	Runs good as of 13 cycles. Still under test
230	Am Cy	9 mg Pt/cm^2	Plat Ni Plaque	20 mg Pt/cm^2	90 KT 10 asb	(1)13.3 (2)13.2	Matrix: Polyethylene membrane Electrolyte: 40.3% KOH; 40 gm (20 gm/matrix). New electrodes	Still under test

*Electrolyte, all cases: 40% KOH; 34 grams by weight unless otherwise noted

Since the ampere-hours used during the discharge cycle decreased with voltage and current falloff, the recharging time also reduced. During the charge periods of the latter cycles the pressure switch would cut off when the pressure reached 350 psi, and the cell would experience a no-charge/no-discharge open-circuit condition for 5 to 10 minutes before the next discharge cycle began. This condition was established to maintain a uniform cycling period of 65 minutes for charging and 35 minutes for discharging. Even with the considerations of degradation and reduced performance observed, this cell test represents the best performance level achieved to date. The 952 cycles at 100 min/cycle are equivalent to 1585 hours (or a period of 66 days) — more than two months of continuous operation.

In examination of the disassembled cell it was not possible to locate the area where the internal short had developed. Since the electrodes were stuck to the matrix, it was necessary to rip part of the electrodes and the matrix in order to separate the cell components. There was black discoloration on the matrix adjacent to each of the electrodes. It appeared to be adhered electrode material that had been ripped away when the electrodes were removed. The internal portions of the matrix were slightly gray in color but showed no substantial difference from matrixes that had been examined after lesser cycling periods. Analysis of the matrix for electrolyte concentration and platinum has not yet been completed, but will be covered during the next period.

Cell 199, put on test during the last period, consisted of an American Cyanamid 9 mg platinum/cm^2 oxygen electrode and a platinized porous nickel plaque hydrogen electrode with a 90 percent KT-10 percent asb matrix. The oxygen electrode used in this cell had been previously used in cell 196 which ran 60 cycles, at which time it developed slow cross gas leakage. The cell was tested continuously for 405 cycles after which the test was discontinued. Figure 2 shows the performance of this cell. As shown, there was a gradual

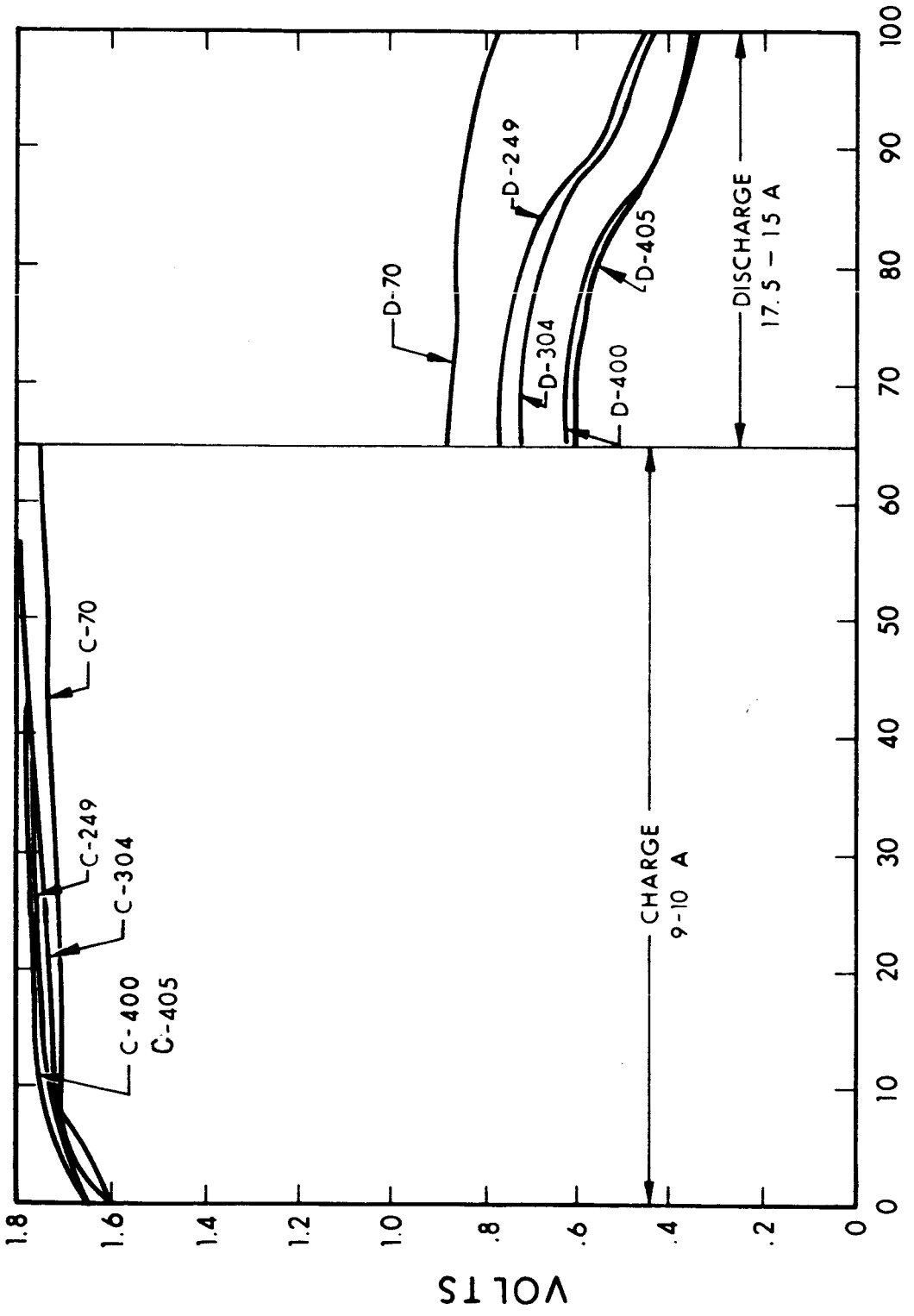

FIG. 2 CYCLING PERFORMANCE OF CELL 199

2614

degradation in performance with cycling. The test was discontinued to examine the internal components, since it appeared no more useful information could be obtained from this cell.

Cell 206 consisted of the same oxygen electrode from cell 199 and a new EOS porous nickel plaque hydrogen electrode with a new matrix composed of 90 percent KT and 10 percent asb. The voltage performance of various cycles of the cell, cycled 700 times, is shown in Fig. 3. As can be seen, there is a gradual increase in the charging voltage with cycling and a gradual decrease in the discharge voltage with cycling. However, the initial performance of the cell showed that the oxygen electrode had not been permanently degraded as a result of its use in the previous cell tests.

Once again the oxygen electrode from cell 206 was reused and assembled in cell 214, which contained a new porous nickel plaque hydrogen electrode with a new matrix of 90 percent KT and 10 percent asb. The hydrogen electrode was a new one since in disassembly the hydrogen electrode of cell 206 was damaged and could not be reused. The cell was cycled continuously for 464 cycles and then developed a slow gas recombination. Figure 4 shows the voltage performance of the cell at various cycles. When the cell was fresh, the performance was as good as the original cells.

Table II summarizes the series of cells employing the same oxygen electrode. A total of 1629 cycles was accumulated with the same oxygen electrode; in every case the performance was equal to its original level.

TABLE II

PERFORMANCE SUMMARY, REUSED OXYGEN ELECTRODE

Cell	Cycles	Test Results
196	60	Developed slow gas recombination
199	405	Gradual degradation in performance
206	700	Gradual degradation in performance
214	464	Developed slow gas recombination
		Total life: 1629 cycles

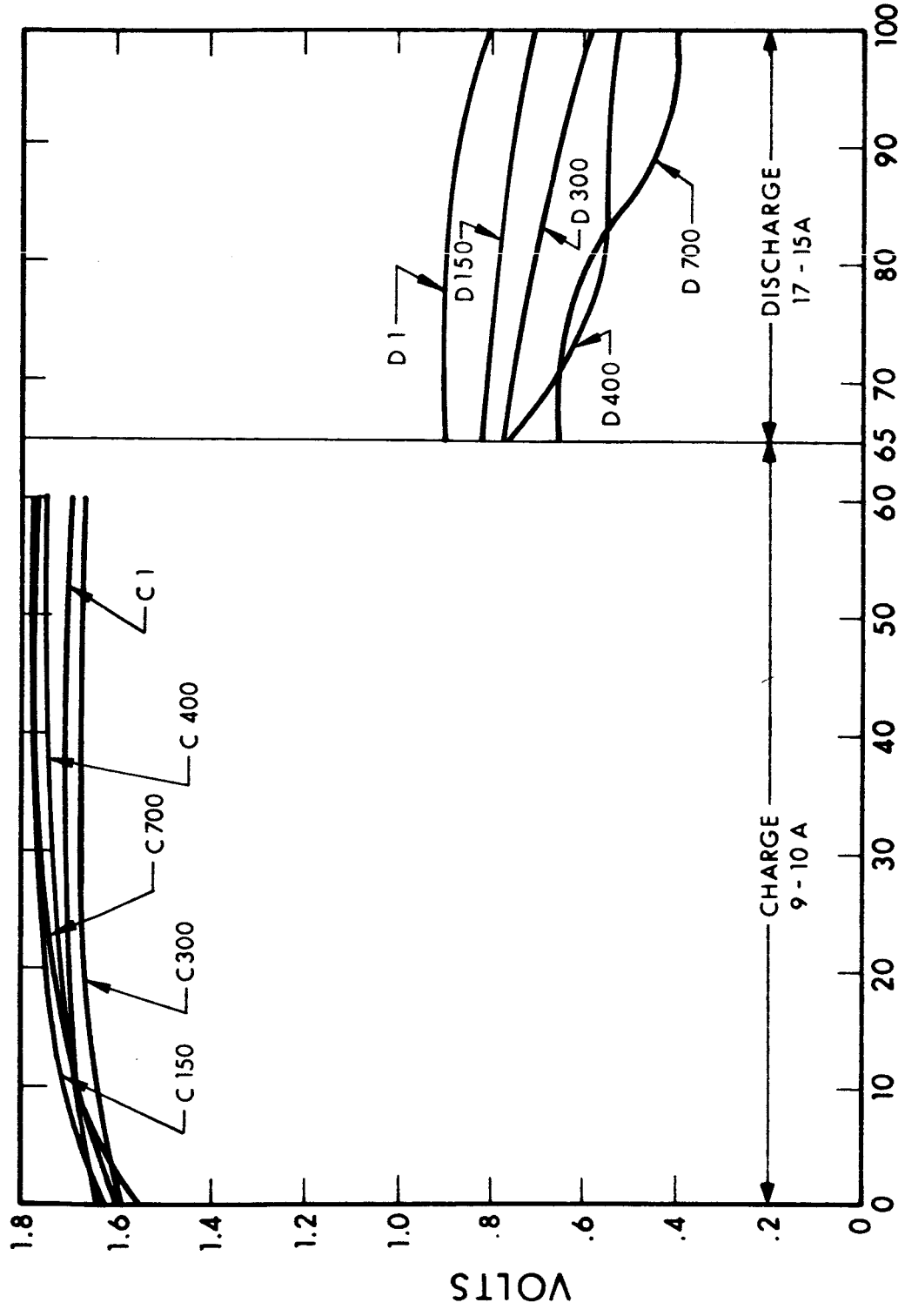

FIG. 3 CYCLING PERFORMANCE OF CELL 206

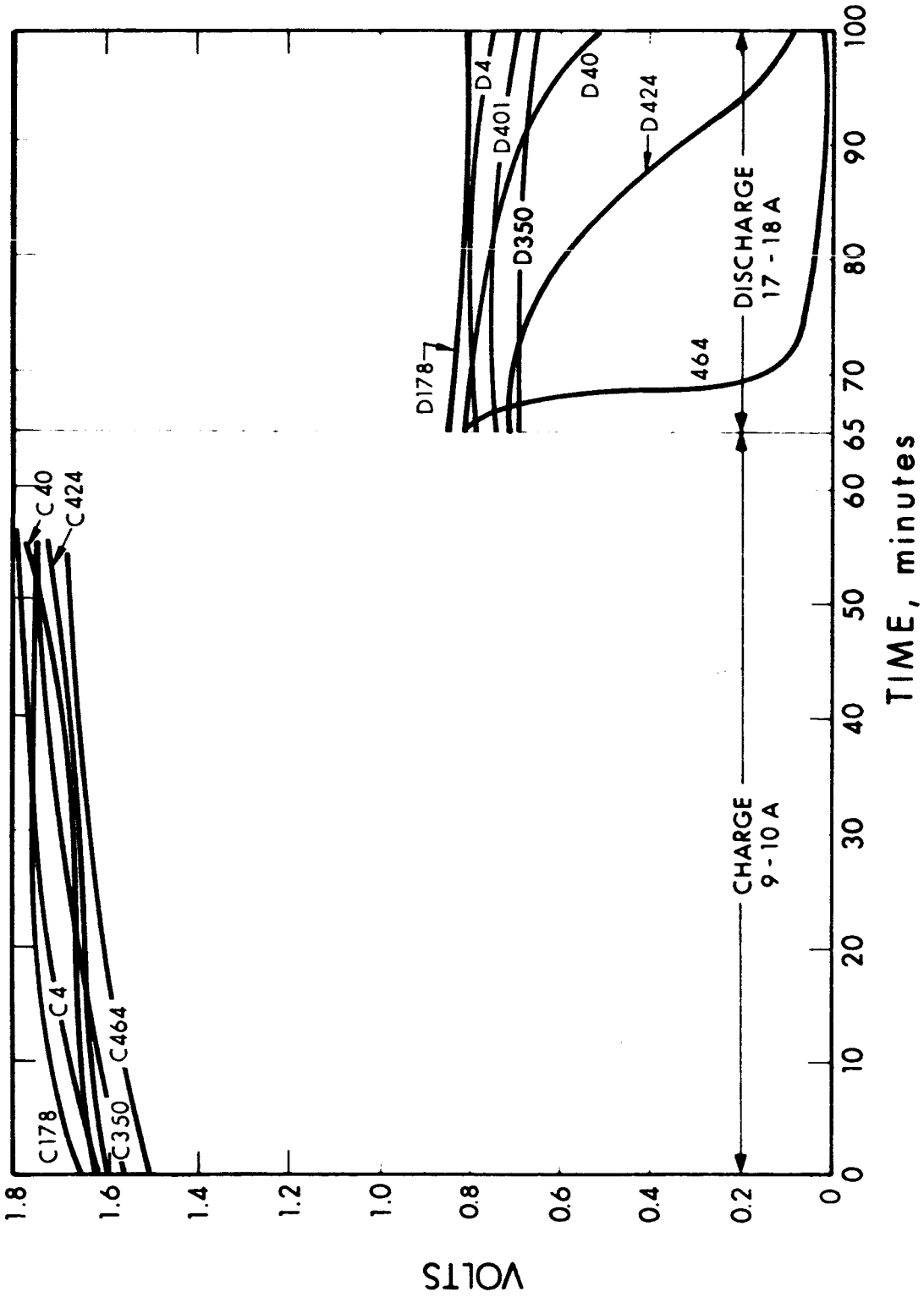

FIG. 4 CYCLING PERFORMANCE OF CELL 214

Cell 202 consisted of an American Cyanamid oxygen electrode which had been previously used in cell 195, and a new porous nickel plaque platinized hydrogen electrode. The cell was subjected to the standard test and operated continuously for 387 cycles, at which time an internal short developed within the cell and the test was discontinued. The cycling performance is shown in Fig. 5. The cell was disassembled and the oxygen electrode was recovered in good condition and reassembled with a new hydrogen electrode and matrix and designated cell 207. This cell showed initial good performance, but developed a slow gas recombination on the 27th cycle and the charge pressure did not rise above 150 psi. The test was then discontinued. The performance of cell 207 is shown in Fig. 6. The oxygen electrode of this cell had also been employed in cell 195. That cell had been subjected to 278 cycles, the results of which are shown in Fig. 7. Therefore, the total accumulated number of cycles subjected to the oxygen electrode was 692 cycles. In each case when the cell was reassembled with a new matrix and hydrogen electrode the initial performance returned to the original level of approximately 0.8 to 0.85V on discharge at 17 to 18A. The results of this and the previous series of tests seemed to indicate that the cause in mode of deterioration encountered with the titanate matrix type cells, now under study, is centered in the matrix or hydrogen electrode and not the oxygen electrode, as previously concluded in asbestos-type cells.

Cell 205 consisted of American Cyanamid oxygen electrode and a platinized nickel plaque hydrogen electrode with a matrix of 90 percent KT, 10 percent asb. The cell was cycled 52 times and showed good initial performance, but at the 53rd cycle, it exhibited a slow recombination, and was unable to recharge the cell above 200 psi; the test was discontinued.

Single cells 208 and 210 contained electrodes that had been previously used in 6-cell units 109 and 110 (described in the eighth quarterly report). Both cells contained matrices of the type used in the 6-cell unit (90 percent KT, 10 percent asb).

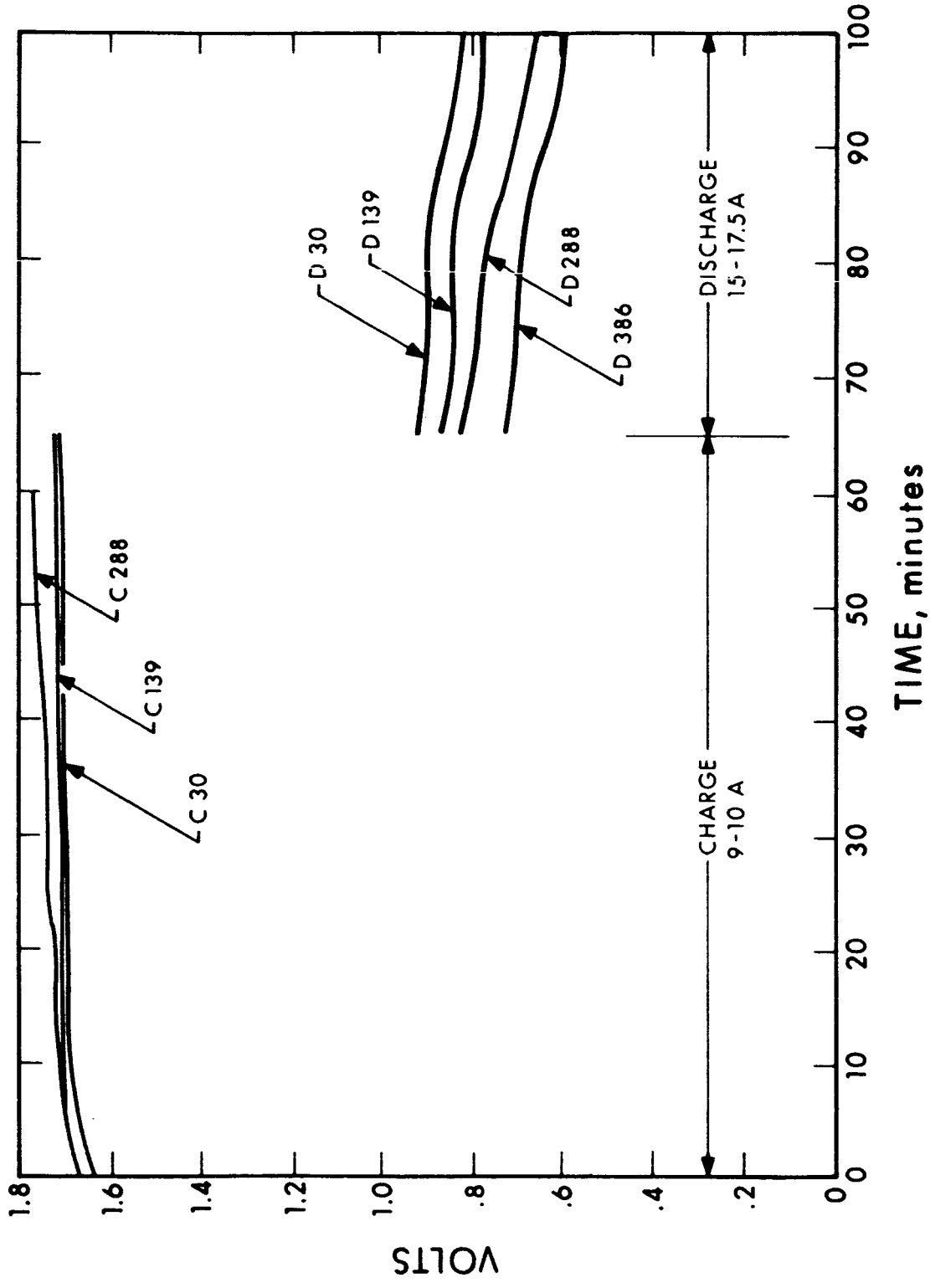

FIG. 5 CYCLING PERFORMANCE OF CELL 202

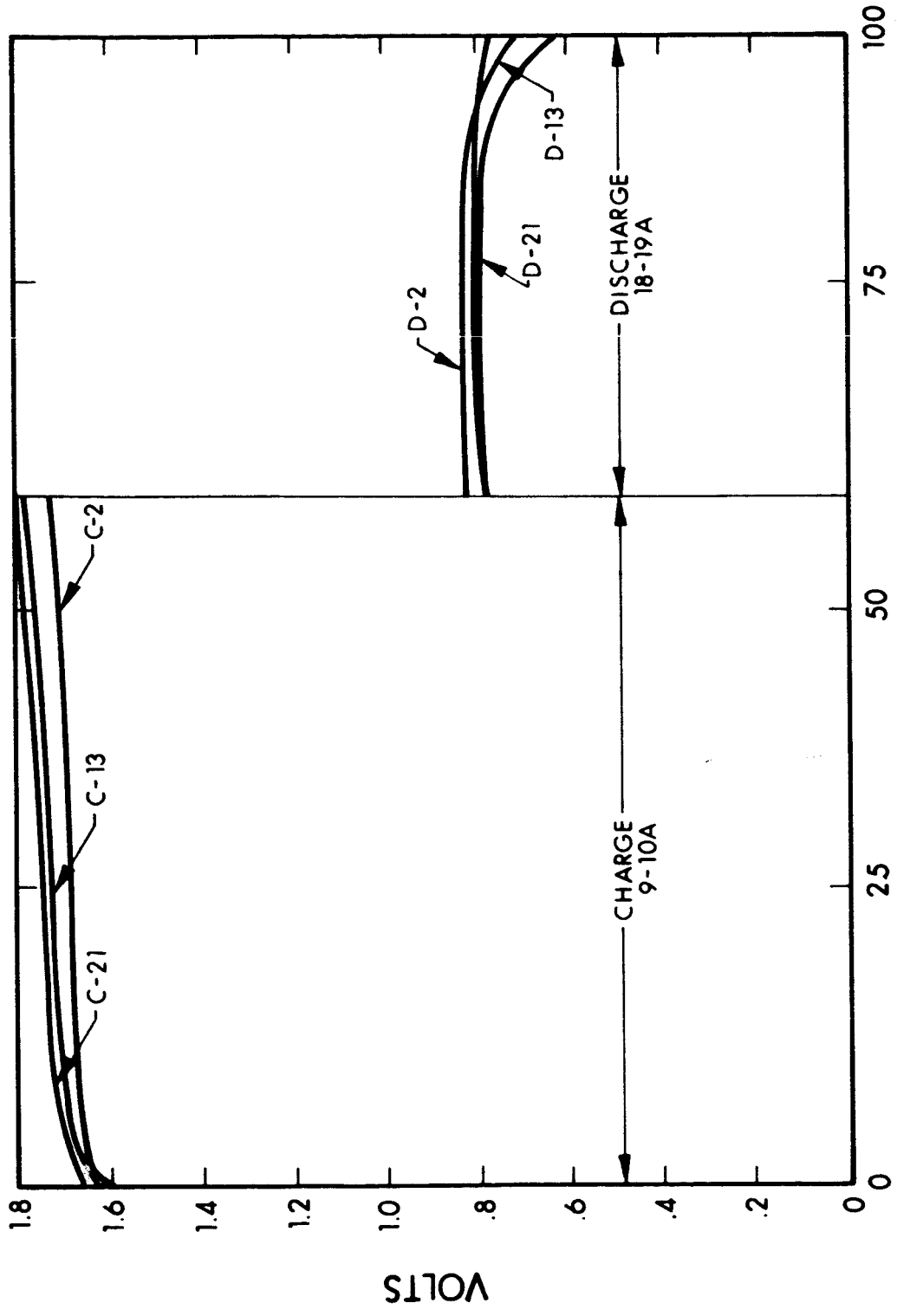

FIG. 6 CYCLING PERFORMANCE OF CELL 207

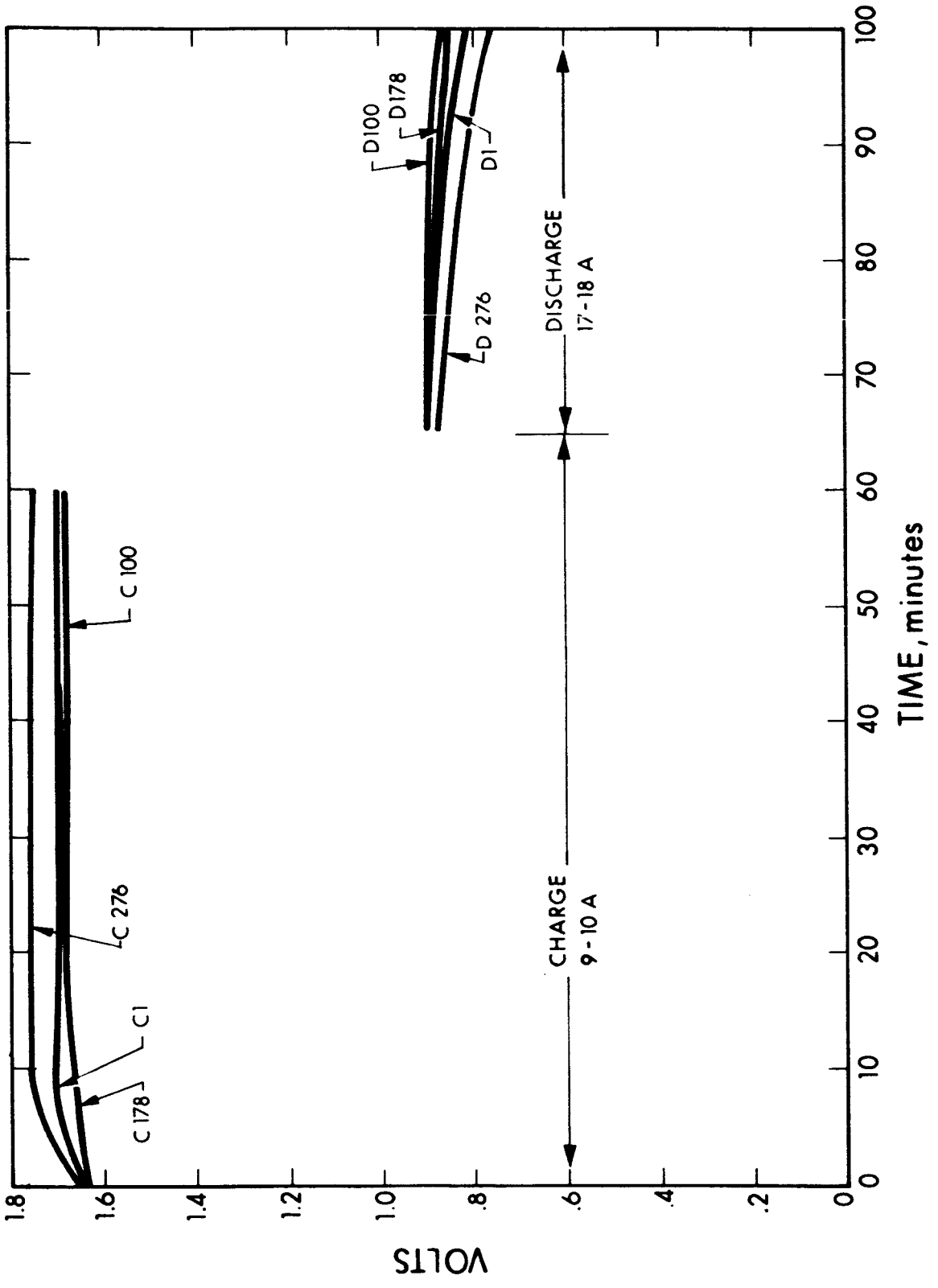

FIG. 7 CYCLING PERFORMANCE OF CELL 195

Cell 208 was cycled for 21 times and showed initial good performance. At that time the test was discontinued.

Cell 210, containing a different set of electrodes, exhibited poor performance by discharging at 0.6 to 0.7V per cell, charging approximately 1.8V per cell. This poor performance of cell 210 confirmed the previous observation on 6-cell testing that the electrodes had somehow been poisoned in the initial testing of 6-cell unit 109, and that was the cause for the poor performance of 6-cell unit 110. Apparently, the only possible poison within the unit as previously observed was the corrosion products of the bipolar plates due to the imperfections in plating of the plates used in 6-cell unit 109.

3.1.2 Ninety Percent KT, 10 Percent Polypropylene Matrix

Cell 211 consisted of an American Cyanamid oxygen electrode and a porous nickel plaque hydrogen electrode with a matrix consisting of 90 percent KT and 10 percent polypropylene fibers. The substitution of polypropylene fibers for asbestos in the matrix was an attempt to eliminate asbestos (which apparently reacts gradually with the electrolyte). The cell was cycled 50 times, showing initial good voltage performance as shown in Fig. 8. Throughout the cycling of the cell, the charge and discharge voltage remained relatively stable; during the last five cycles, however, a slow gas recombination took place within the cell such that the cell pressure did not reach the fully charged level. On subsequent discharges there was a falloff in the discharge voltage. The slow gas recombination that occurred in the cell was apparently due to the structural weakness and imperfection of the matrix; thus, no conclusion could be drawn as to the ability of this type of matrix to enhance cycle life of the cell.

Cells 213 and 215 were additional attempts to use a matrix of 10 percent polypropylene fibers. In both cases, the oxygen electrodes were American Cyanamid types and the hydrogen electrodes

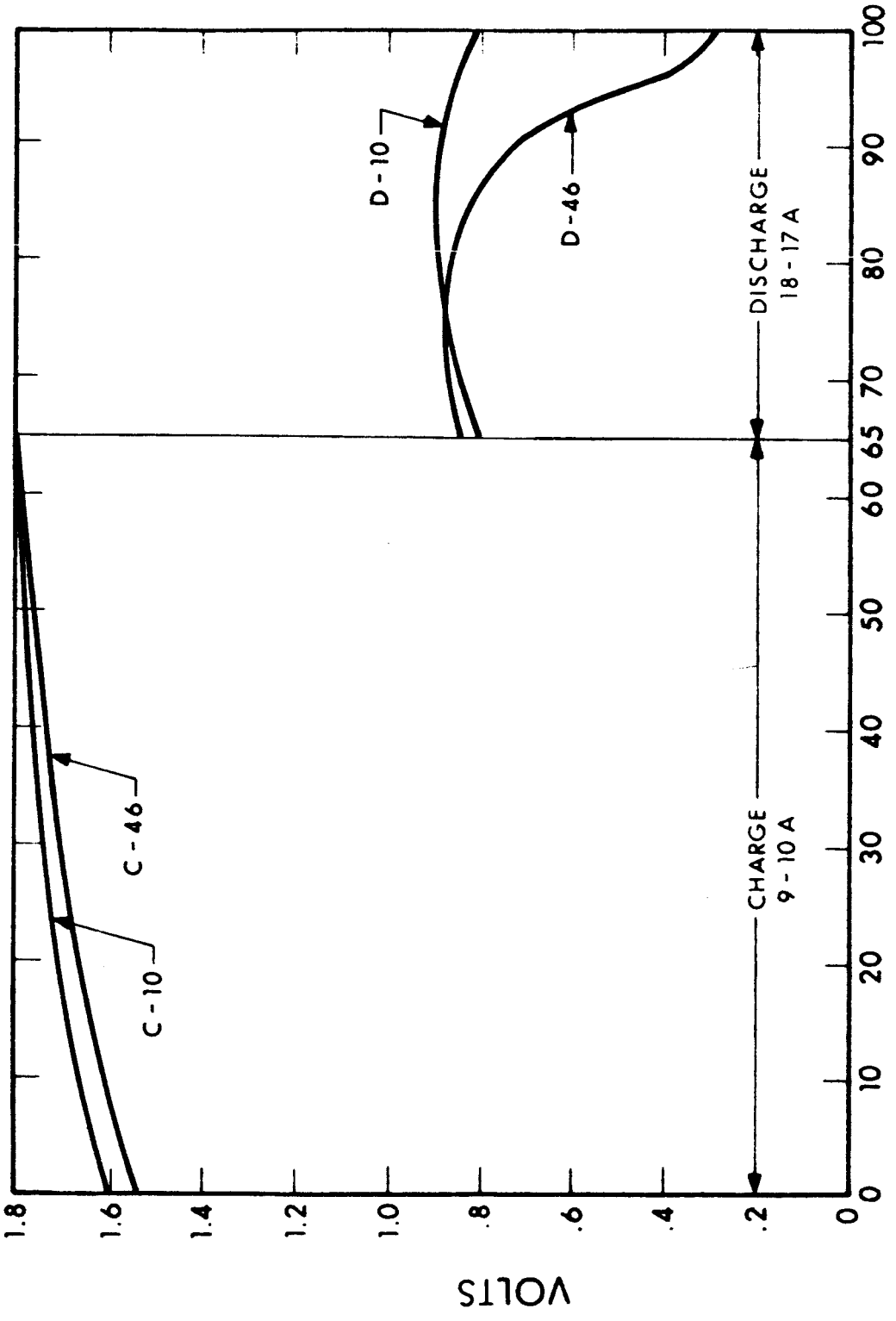

FIG. 8 CYCLING PERFORMANCE OF CELL 211

were platinized nickel porous plaques. Cell 213 was cycled 82 times with relatively stable voltage performance, as shown in Fig. 9. Beyond that point, a slow gas recombination during the charge cycle took place and the cell did not reach a full state of charge. On subsequent discharges there was a rapid falloff in discharge voltage as the cell pressure dropped to levels of 50 psi and below. Cell 215 showed initial good performance, but on the 30th cycle it also developed a slow gas recombination, resulting in a falloff in the discharge voltage as the pressure decreased below 50 psi.

Cells 223 and 224 were the final attempts with a matrix of 10 percent polypropylene fibers. Each of the cells was assembled with an American Cyanamid oxygen electrode and porous nickel plaque hydrogen electrodes. Cell 223 developed a slow gas recombination on the 22nd cycle and cell 224 developed a slow gas recombination on the 24th cycle.

The performance with the 10 percent polypropylene matrixes of the five cells resulted in a gradual slow recombination after a period of cycling. The method of fabricating these matrixes was apparently inadequate, and the resulting matrixes had flaws or pinholes that developed with cycling.

From the results obtained, it is apparent that this type of structure is not capable of withstanding cross gas leakage. Different techniques will have to be employed to fabricate chemically inert matrixes without asbestos constituents.

3.1.3 Hydrogen Concentration Cell Tests

Based on some of the previous results indicating that modes of degradation now encountered are caused by the matrix and/or hydrogen electrode, a new hydrogen concentration cell test was set up and designated cell 209. In this case, two platinized porous nickel plaque electrodes were employed. The matrix was 90 percent KT and 10 percent asb. The cell was run in a continuous concentration mode at 18A (equivalent to 100 mA/cm^2). Figure 10 shows the voltage performance of this cell as a function of time.

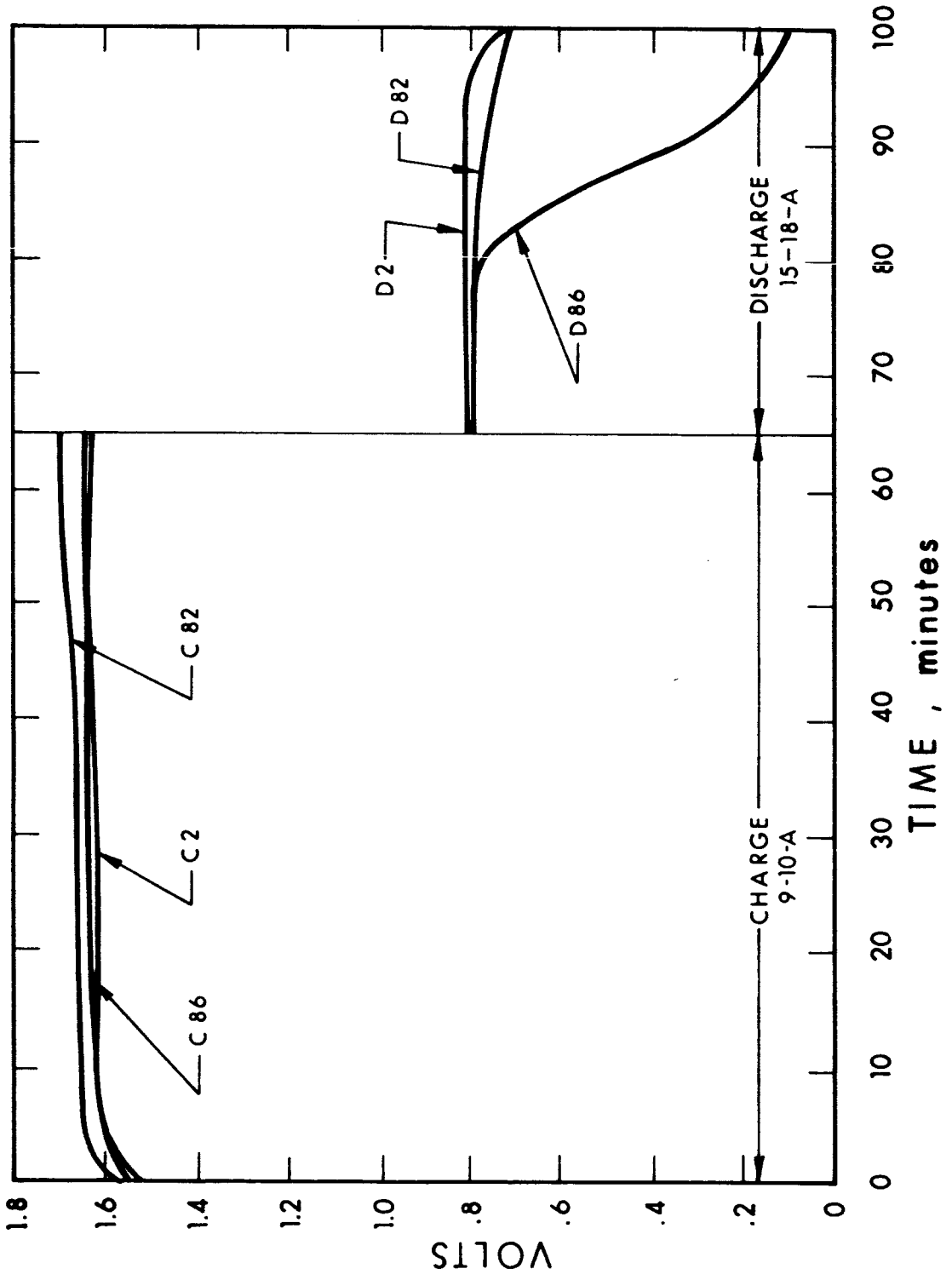

FIG. 9 CYCLING PERFORMANCE OF CELL 213

360

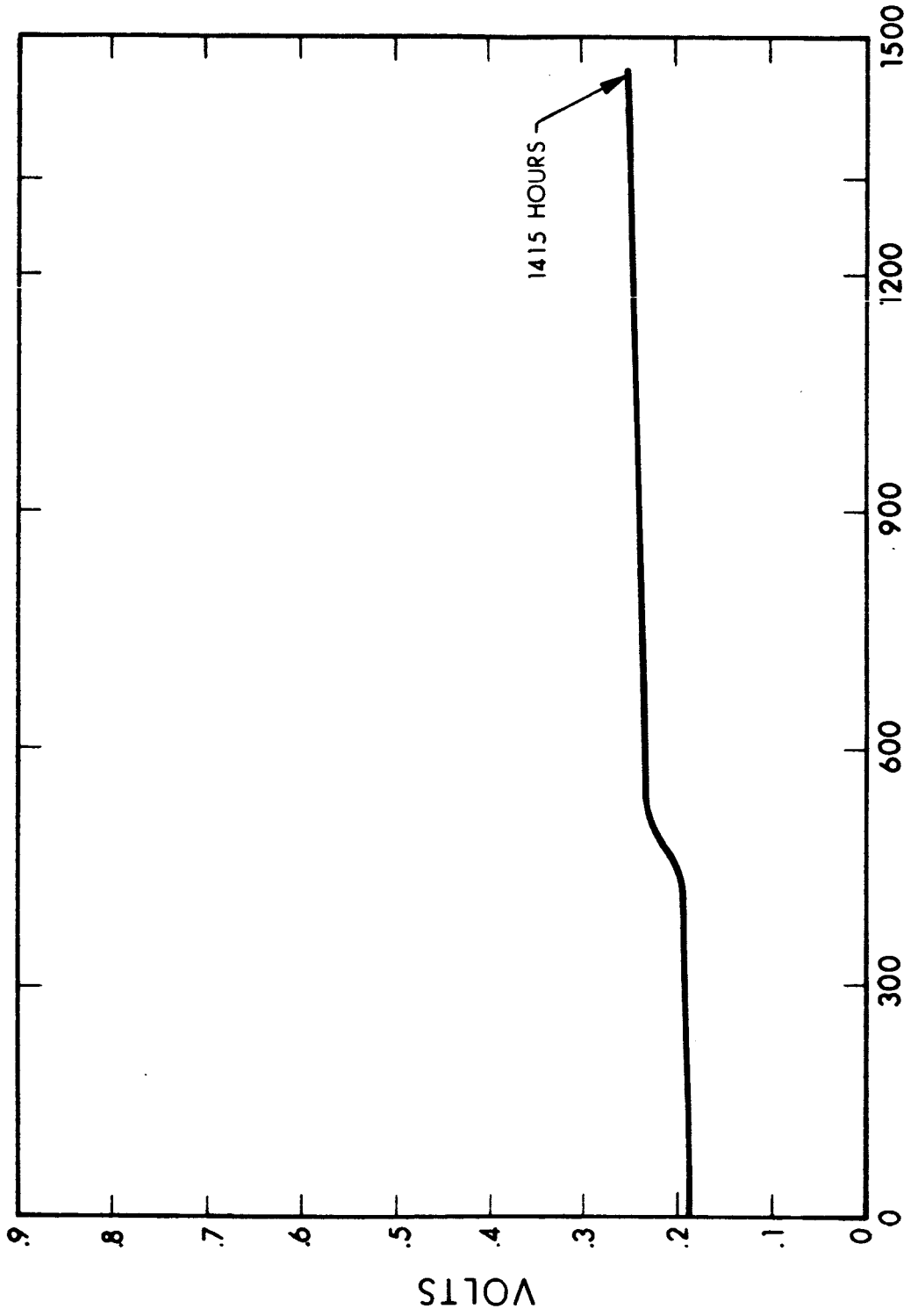

FIG. 10 H_2 CONCENTRATION, CELL 202

As can be seen, at approximately the 500-hour point there was an increase in the voltage which leveled off again and remained relatively flat to the 1415-hour point where the test was discontinued. This test seems to indicate that the hydrogen electrodes are not substantially affected by long-term use in the concentration mode.

Cell 218 was a repeat of the hydrogen concentration cell test. Both electrodes consisted of platinized porous nickel plaques and the matrix was made of 90 percent KT and 10 percent asb. The construction of the cell was identical to that of cell 209. The cell was put on a continuous-current load of 18A equivalent to 100 mA/cm^2. The results of the cell are shown in Fig. 11. To date the cell has achieved in excess of 600 hours of life and is still under test. The voltage of the cell is stable at approximately 0.1V which is lower than the voltage achieved with cell 209. The results of the two concentration cells show that there is a very slow negligible deterioration of the hydrogen electrodes when they are subjected continuously either to the charge or discharge mode. In addition, if an electrolyte reaction is occurring with the matrix and the electrolyte, this reaction is not affecting the performance of the hydrogen electrodes.

3.1.4 Membrane Cell

Many of the cell tests employing the 10 percent asbestos or polypropylene matrixes resulted in the development of cross gas leakage as cycling proceeded. This cross gas leakage apparently occurs due to structural changes in the matrix or imperfections resulting from the processing techniques employed. To overcome this deficiency, an alternate approach is being considered, which involves use of a thin membrane material that would be located between two layers of an absorbent matrix. This "sandwich" is capable of storing and holding a large quantity of electrolyte yet is resistant to cross

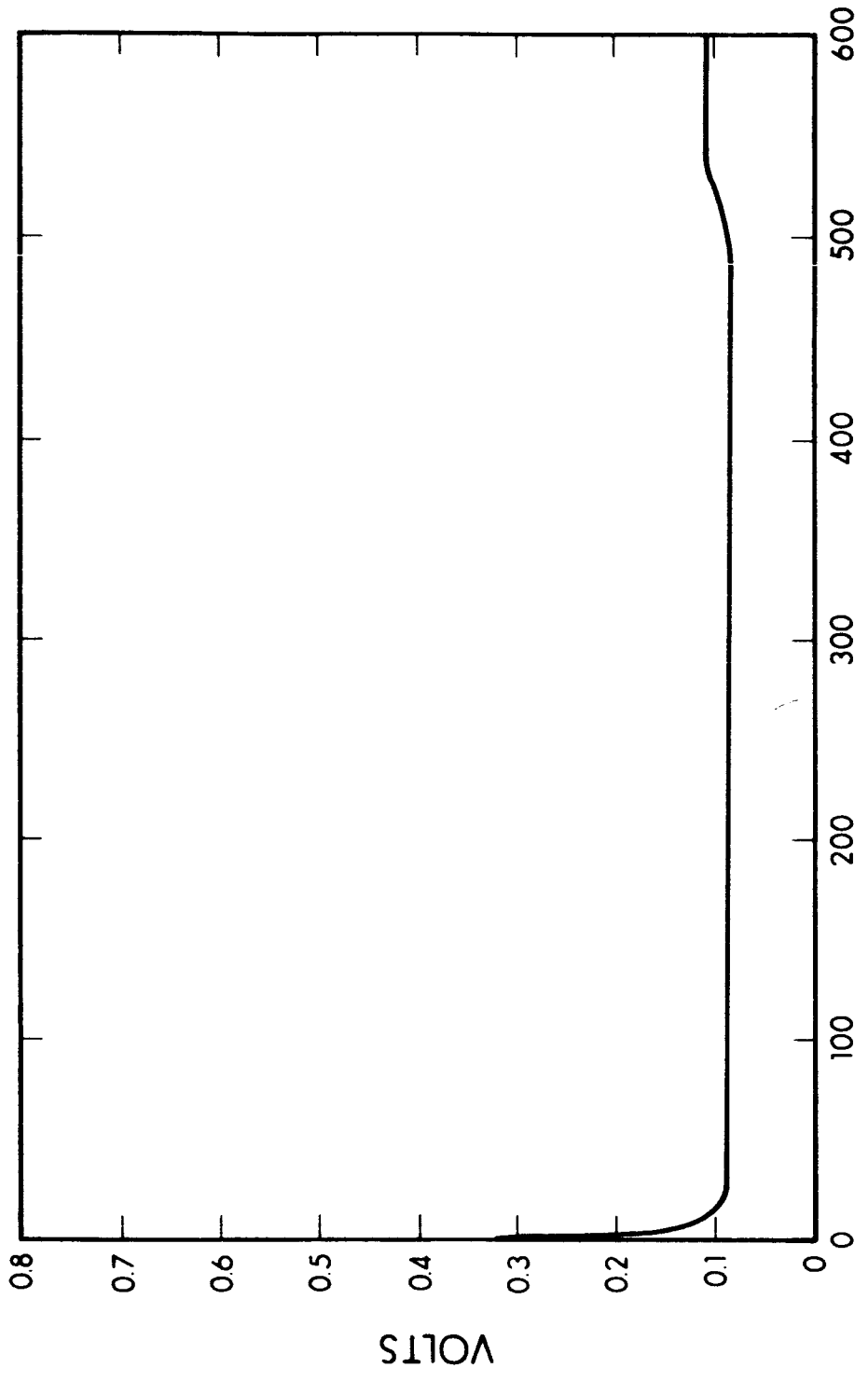

FIG. 11 H_2 CONCENTRATION, CELL 218

gas leakage. Since the absorbent matrix in this configuration would not have to prevent the cross gas leakage, it would be possible to reduce matrix compression and increase the electrolyte quantity, thereby possibly increasing performance and capacity.

Cell 222 was the first attempt at such a configuration. It consisted of a membrane of radiated polyethylene (permion 300) normally employed in conventional secondary batteries and a layer of 90 percent KT and 10 percent asb on either side of the membrane. The compression on the membrane was reduced by eliminating the backup screens behind the gas electrodes and 25 grams of electrolyte was impregnated in each half of the matrix. The cell was put on test and subjected to five cycles. The test was discontinued because the cell exhibited very poor performance.

Cell 226 was similar in construction to cell 222 and consisted of a sandwich matrix of a radiated polyethylene membrane, and two layers of the KT with asb. However, in this case, the backup screens were used behind the electrodes and the electrolyte quantity was reduced to 20 gm per matrix. The cell was put on test and subjected to four cycles; the test was discontinued when the cell showed poor performance.

Cell 227 was similar in construction to cell 226 except that a 0.05-inch spacer between the electrodes was used instead of a 0.04-inch type normally used in the cell assemblies. This cell was damaged due to an excessive differential pressure that occurred in the flushing, and the cell was never subjected to test.

Cell 228 consisted of a membrane made of woven Teflon cloth between two layers of KT with 10 percent asb. The cell was cycled 39 times but exhibited poor performance. Apparently, the nonwetting nature of the woven Teflon material resulted in a high cell impedance.

Cell 229 consisted of an American Cyanamid type oxygen electrode, a porous nickel plaque hydrogen electrode, and a

matrix that was made of a sandwich of two layers of KT and 10 percent asb with a cellulose membrane, designated FSC (also employed in conventional batteries). It was realized that this type of membrane would not withstand the high concentration electrolyte elevated temperature conditions in the cell very long, but it was felt that for feasibility this test should be conducted. The cell was assembled with a 0.05-inch spacer, and 20 grams of electrolyte in each of the two matrix halves. The cell was put on test and showed good performance. The results are shown in Fig. 12. The cell has achieved 13 cycles to date, and performs similarly to cells that did not contain membranes, demonstrating that the membrane approach is feasible and does not cause excessive increases in internal impedance.

Cell 230 consisted of an American Cyanamid type oxygen electrode, a porous nickel hydrogen electrode, with a sandwich matrix consisting of radiated polyethylene membrane and two layers of KT with 10 percent asb. This cell also contained 20 grams of electrolyte per matrix with a 0.05-inch spacer similar to cell 229. The first cycle of this cell showed good performance, and the cell is still on test.

The use of a membrane configuration offers several potential significant advantages: (1) cross gas leakage can virtually be eliminated, (2) it increases the scope of types of matrix materials that can be used for electrolyte absorbency, and (3) it reduces the stringent physical requirements on a matrix. With regard to the third advantage, matrixes inherently must have a characteristic of high quantity of electrolyte absorption, and so must be somewhat porous, yet they should be nonporous to the extent of preventing cross gas leakage.

Tests have demonstrated the feasibility of a membrane sandwich cell by virtue of the satisfactory performance of such a cell. It is felt that the approach warrants further effort to determine the correct matrix-membrane electrolyte compression ratio to obtain stable performance.

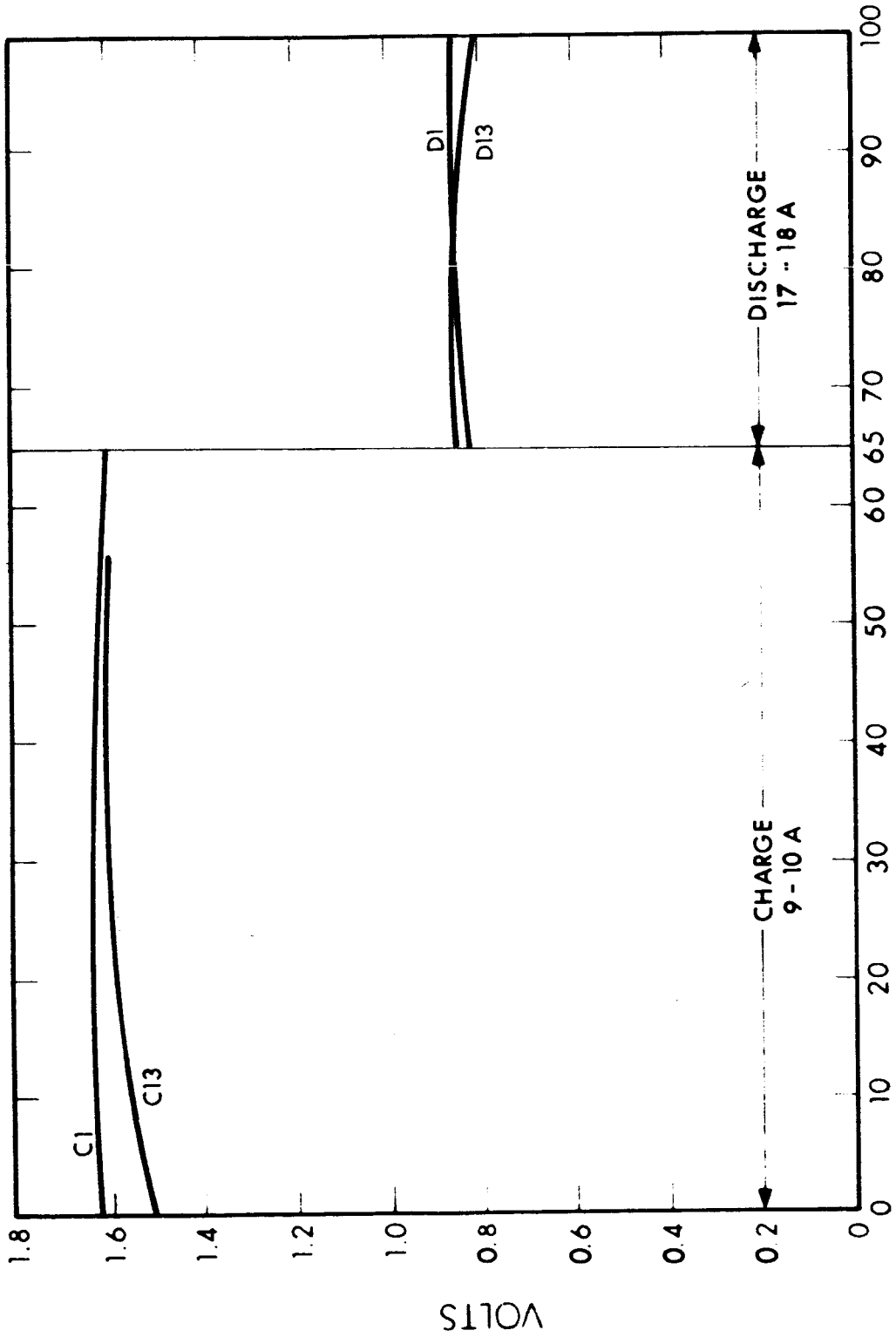

FIG. 12 CYCLING PERFORMANCE OF MEMBRANE SANDWICH CELL 229

3.1.5 Other Single Cell Tests

Cell 212 consisted of an American Cyanamid oxygen electrode containing platinum of 40 mg/cm^2 (as opposed to the standard type electrodes which contain 9 mg/cm^2). The matrix was 90 percent KT and 10 percent asb; the hydrogen electrode was the standard nickel porous plaque type. The cell was subjected to 418 charge/discharge cycles. The voltage performance at various cycles is shown in Fig. 13. As can be seen, there was a gradual degradation in performance with cycling. In addition, this cell exhibited the slow gas recombination which resulted in a rapid falloff in the voltage performance during discharge in the latter cycles. The electrodes were removed from this cell unwashed and preassembled with a new matrix consisting of the same type. This new cell (designated 217) was cycled continuously for 432 cycles to a point of slow gas recombination.

The results shown in Fig. 14 indicate that an American Cyanamid electrode of 40 mg. Pt/cm^2 has the same performance characteristics as the 9 mg Pt/cm^2 electrodes normally used.

Cell 216 consisted of the standard 9 mg. Pt/cm^2 American Cyanamid electrodes for both the hydrogen and oxygen sides and a matrix of 90 percent KT, 10 percent asb. The American Cyanamid electrode was employed on the hydrogen side to see if its use would improve performance and life. The cycling results are shown in Fig. 15. As can be seen, a gradual deterioration of charge and discharge voltage was observed. This rate of degradation was greater than that normally obtained with the nickel plaque hydrogen electrode cells.

Cell 219 consisted of an American Cyanamid oxygen electrode, a porous nickel plaque hydrogen electrode, and a 90 percent KT, 10 percent asb matrix. The spacing between the electrodes was increased by removing the gas backup screens and an excess of electrolyte was added to the matrix. This test was an attempt to see if increasing the electrolyte quantity and reducing the compression would

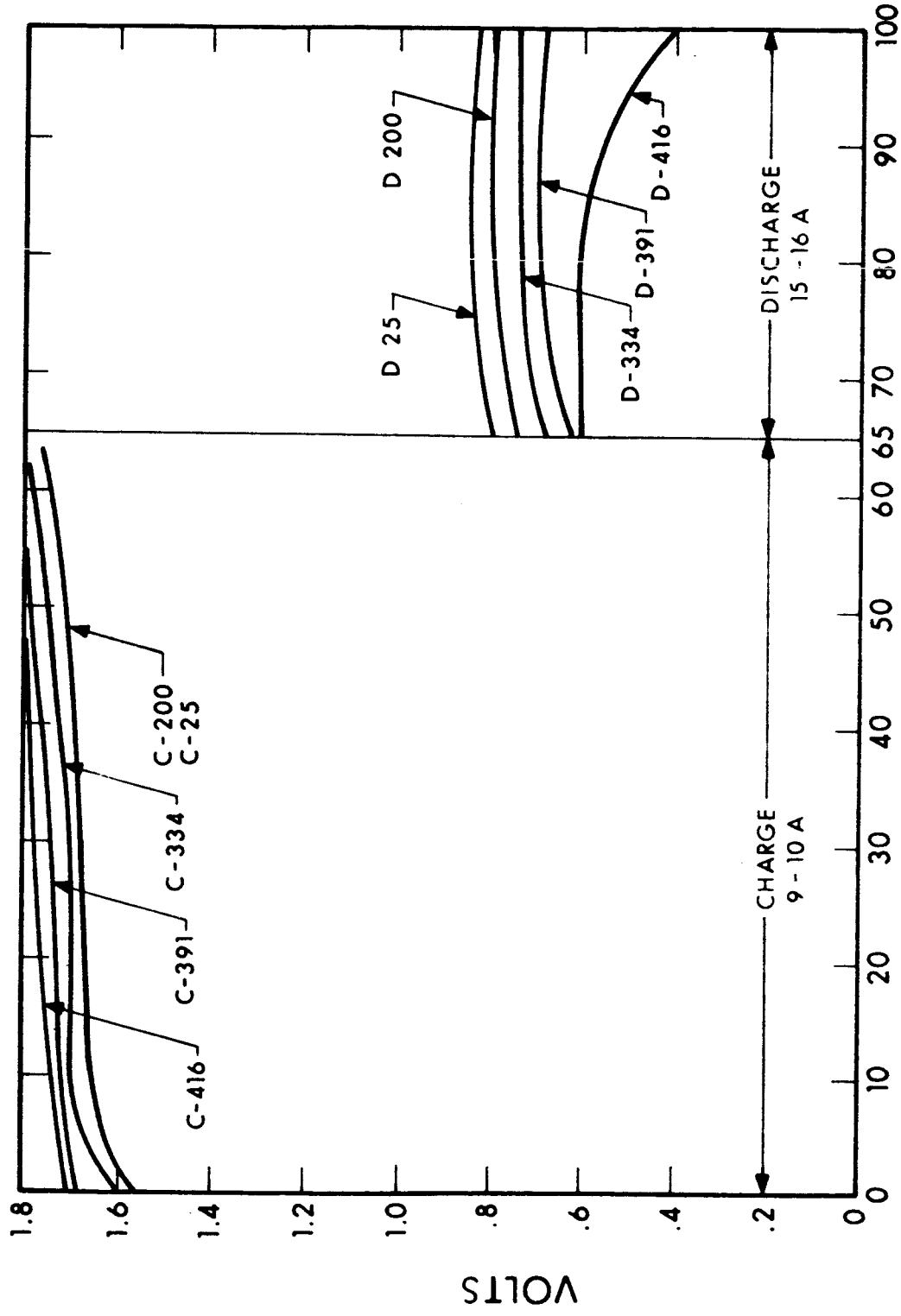

FIG. 13 CYCLING PERFORMANCE OF CELL 212

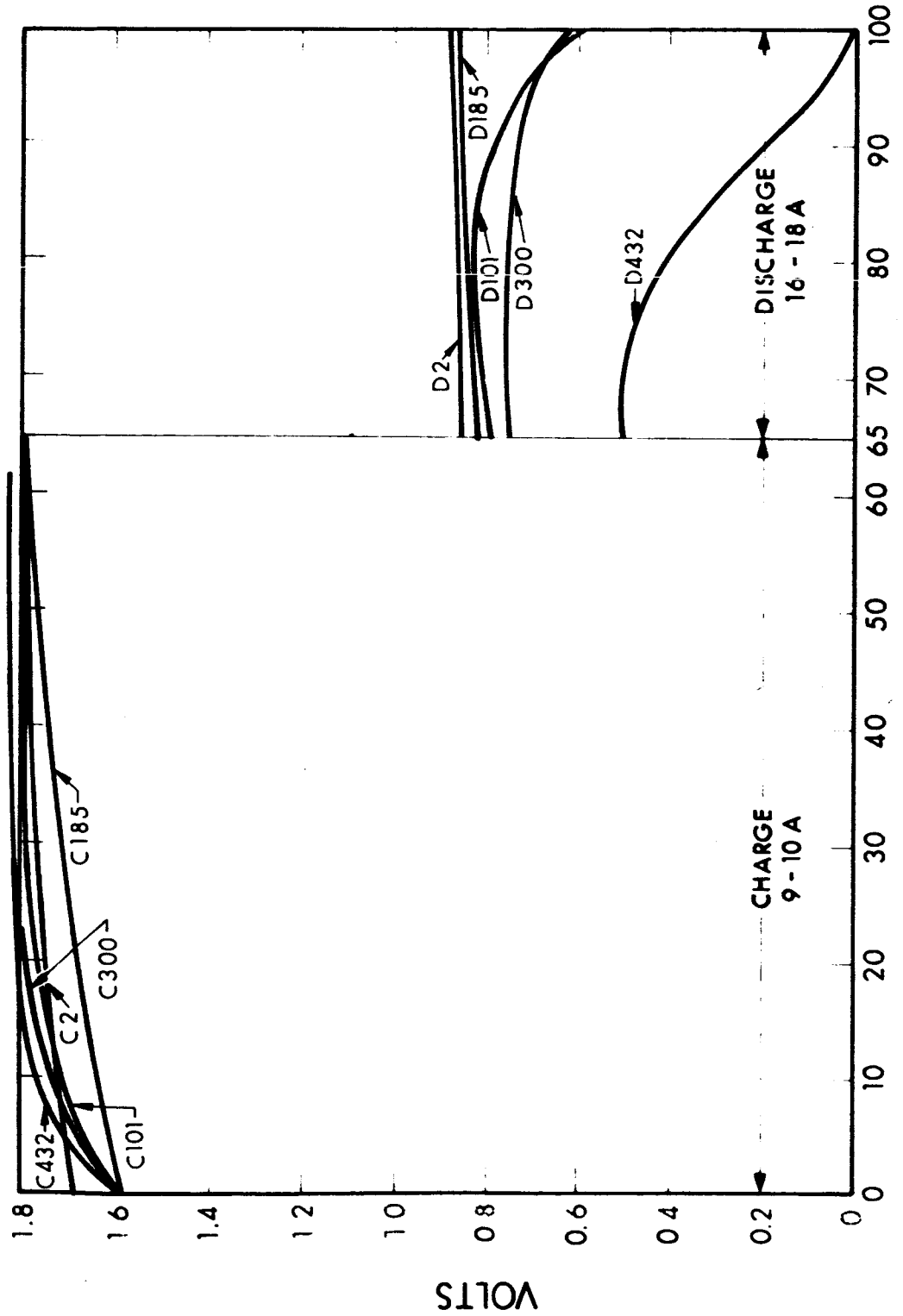

FIG. 14 CYCLING PERFORMANCE OF CELL 217

4110-QL-9

3735

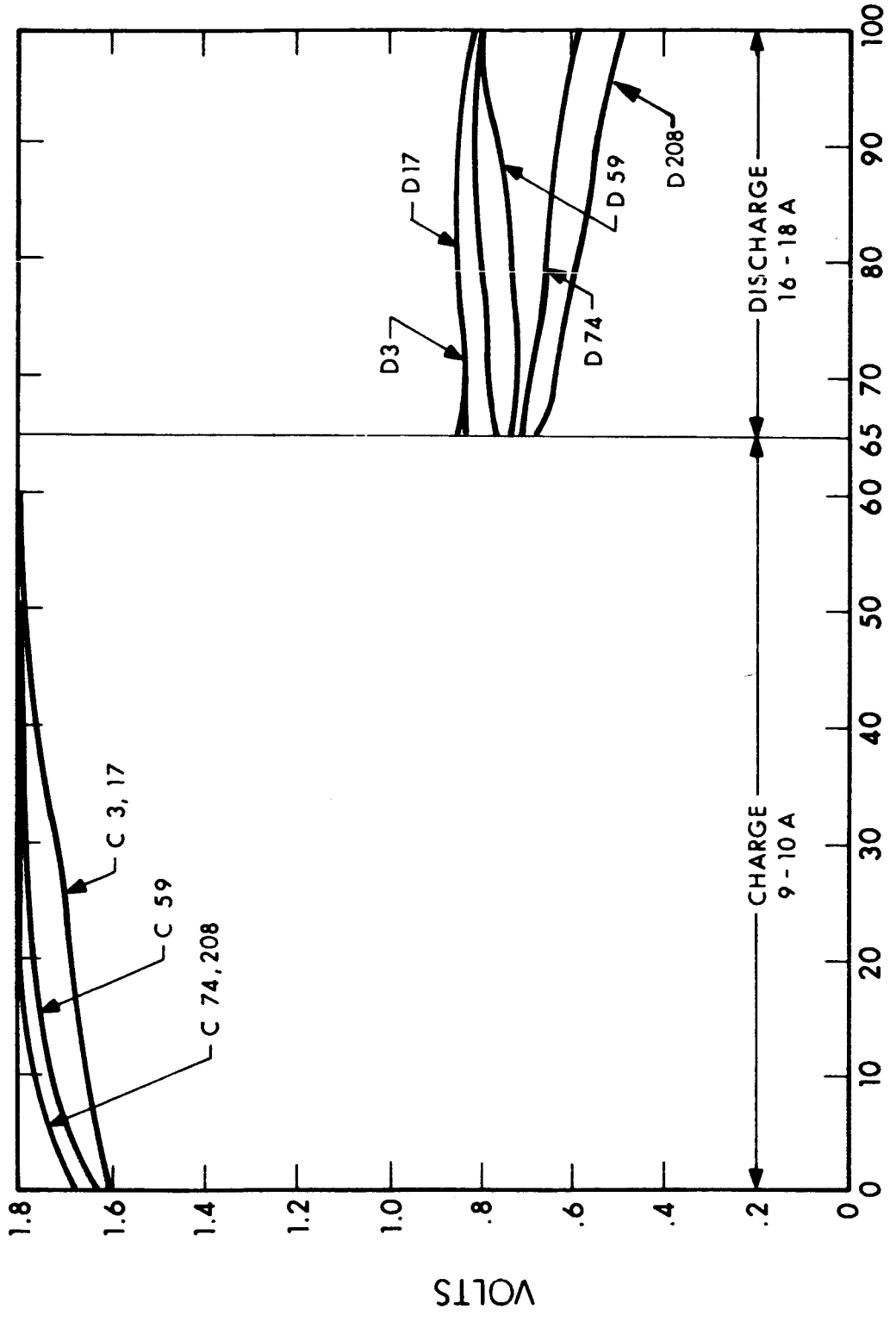

FIG. 15 CYCLING PERFORMANCE OF CELL 216

improve performance and reduce cross gas leakage. The cell showed cross gas leakage after 85 cycles, indicating that this approach is not satisfactory. To further evaluate this type of cell construction, two additional cells (220 and 221) were assembled, using electrodes identical with the types of cell 219. Standard fuel cell grade asbestos (0.060 inch thick) was used as the matrix since it was felt that this matrix material was more uniform than the titanate matrixes and would thus prevent cross gas leakage. Once again, 50 grams of 40 percent electrolyte was impregnated in the matrix. Cell 220 exhibited a high internal impedance indicating that there was poor electrode-electrolyte contact due to the large spacing between electrodes; the cell was not subjected to test. Cell 221 was subjected to 15 cycles at which time it developed cross gas leakage. Considering the results of the previous tests, this electrode matrix configuration doesn't appear to offer any promise of improvement.

3.2 Multicell Testing

Based on the promising performance in single cells using the American Cyanamid oxygen electrode, platinized nickel plaque hydrogen electrode configuration, and a matrix of KT with 10 percent asb, a new 6-cell unit was built during this period to be subjected to test. During checkout of the cell stack, the unit is subjected to slight differential pressure to determine if any cross leakage exists. In this checkout, it was found that there was an appreciable cross gas leakage through the matrixes which would have resulted in a slow recombination within the stack; the unit was therefore disassembled. An examination of the matrixes revealed no obvious area where the cross leakage was occurring, except that many of the matrixes were found to be thinner in the center than on the edges. A review of the process technique of the matrixes revealed that a slightly convex Buchner funnel was utilized in the filtrate formation of the matrixes which would result in less material being in the center of the matrix.

To alleviate this problem, a new funnel that has a flat inner plate will be used for fabrication of future matrixes. A new series will be fabricated and the 6-cell unit reassembled, checked out, and subjected to test in the next period.

3.3 500-Watt 34-Cell Unit

In preparation for the ultimate delivery of a 34-cell 500W unit, a new set of tankage was ordered. The new tanks, which contained a smaller flange to reduce weight, were fabricated of aluminum and nickel-plated to protect the aluminum surface. These tanks were delivered during this period.

3.4 Potassium Titanate Analysis

In order to obtain a better understanding of the composition and impurities in the pigmentary KT used in the matrixes, a sample was submitted to an outside testing laboratory for spectrographic analysis; the results are shown in Table III.

TABLE III

SPECTROGRAPHIC ANALYSIS OF PIGMENTARY POTASSIUM TITANATE

Titanium	47%	Aluminum	0.071%	Calcium	0.11%
Potassium	17%	Silicon	0.042%	Lead - less than	0.02%
Magnesium	0.020%	Other elements - nil			

This analysis confirmed roughly the composition of KT as supplied by DuPont.

4. CONCLUSION

From the results of single cells tested in this period, it appears that the oxygen electrodes are not affected or degraded by cycling with the potassium-asbestos matrixes over long periods of accumulated cycles. However, there is unquestionably a gradual degradation in performance with cycling that could be attributed to the hydrogen electrode or the matrix electrolyte composition. From the performance of the hydrogen concentration cell it appears that degradation is not due to the hydrogen electrode; it is most probably due to a matrix electrolyte reaction. For structural considerations, all matrixes with long-life cells have contained 10 percent asbestos; it is possible that this asbestos could be causing the problem. Attempts to substitute polypropylene for the asbestos fibers have not proved successful due to the structural weakness of these matrixes and the resultant cross gas leakage.

The approach of mating a sandwich matrix with a gas barrier membrane relaxes the structural requirements of the absorbent matrix. This will enable the use of pure potassium titanate matrixes and other materials that normally would not prevent cross gas leakage on their own.

Even with the degradations and problems observed, the use of matrixes of KT with asbestos enables the achievement of reasonably consistent lifetimes in excess of 500 cycles. One such cell has achieved 952 cycles.

5. PLANS FOR THE NEXT PERIOD

Cycling single-cell tests will be continued to evaluate KT matrixes of different total weight, electrolyte-to-matrix weight ratio, thickness, compression ratio, and additions of asbestos, Teflon, and other fibers that can improve the structure of the matrix. Tests will be continued to determine reaction and corrosion rate of KT with KOH. Analytical tests of matrixes removed from cycling cells will be conducted to determine if platinum migrations exist within the matrix and if electrolyte reductions and consumption occur during cell cycling. Additional membrane-sandwich matrixes will be evaluated with different membrane-matrix electrolyte configurations. Six-cell units employing promising electrode-matrix configurations will be assembled and subjected to life testing to obtain additional experience and test data of multicell units. When stable reproducible performance is obtained with 6-cell units, a cell configuration will be frozen and testing of the 34-cell 600-W unit will be initiated.